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For: Eastman Chemical Company

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(54) [Title of the Invention] **Polyester Elastomer Composition for Calendering**

(57) [Summary]

**[Object]** The present invention relates to a polyester elastomer composition capable of yielding sheets or films that have adequate calendering properties, particularly plate-out resistance, as well as excellent surface appearance, mechanical properties, heat resistance during processing, and color tone during processing.

**[Composition]** A polyester elastomer composition for calendering, obtained by melting and admixing a phosphorus-based compound into a polyester block copolymer containing as principal constituent components thereof (A) high-melting crystalline polymer segments and (B) low-melting polymer segments comprising aliphatic polyether units and/or aliphatic polyester units.

**[Claims]**

**[Claim 1]** A polyester elastomer composition for calendering, obtained by melting and admixing a phosphorus-based compound in an amount of 0.001–10 weight parts per 100 weight parts of a polyester block copolymer containing as principal constituent components thereof (A) high-melting crystalline polymer segments and (B) low-melting polymer segments comprising aliphatic polyether units and/or aliphatic polyester units.

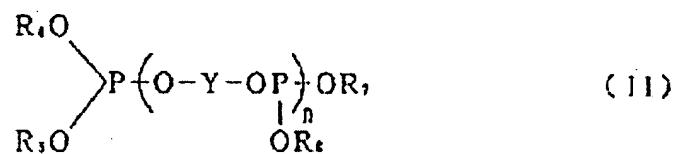
**[Claim 2]** The polyester elastomer composition according to claim 1, wherein the phosphorus-based compound is one or more compounds selected from the compounds expressed by General Formulas (I), (II), and (III).

**[Chemical Formula 1]**

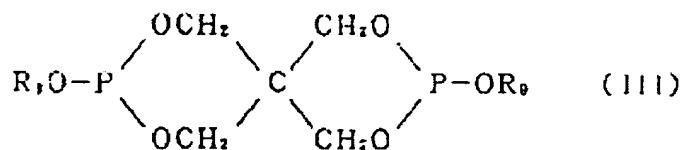


(In Formula (I), R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each a hydrogen atom; a group containing a C<sub>1-60</sub> substituted or unsubstituted alkyl, alkenyl, aryl, polyoxyalkylene alkyl ether, or polyoxyalkylene aryl ether; or one of these substituted groups; wherein the R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> may be bonded to each other with or without the aid of substituents.)

[Chemical Formula 2]



[Chemical Formula 3]



(In Formulas (II) and (III),  $\text{R}_4$ – $\text{R}_9$ , which may be the same or different, are each a group containing hydrogen, alkyl, aryl, cycloalkyl aryl, alkyl aryl, aryl alkyl, alkenyl, aryl alkenyl, or ether bonds; one of these groups containing an epoxy; or one of these substituted groups.  $\text{Y}$  is a polyhydric alcohol or polyhydric phenol residue.  $n$  is 0 to 10.)

**[Claim 3]** The polyester elastomer composition according to claim 2, wherein the phosphorus-based compound is a metal salt containing the compound of claim 2 as a counterion.

**[Claim 4]** A polyester elastomer composition for calendering, obtained by melting and admixing a phosphorus-based compound in an amount of 0.001–10 weight parts and a polyolefin wax in an amount of 0.001–10 weight parts per 100 weight parts of a polyester block copolymer containing as principal constituent components thereof (A) high-melting crystalline polymer segments and (B) low-melting polymer segments comprising aliphatic polyether units and/or aliphatic polyester units.

#### **[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a polyester elastomer composition capable of yielding sheets or films that have adequate calendering properties,

particularly plate-out resistance, as well as excellent surface appearance, mechanical properties, heat resistance during processing, and color tone during processing.

[0002]

**[Prior Art]** Polyester block copolymers in which crystalline aromatic polyester units serve as hard segments (and aliphatic polyether units such as poly(alkylene oxide)glycols, or aliphatic polyester units such as polylactones serve as soft segments) have excellent shock resistance, elastic recovery, flexibility, and other mechanical properties in addition to superior high-temperature characteristics, adhesion, and numerous other properties, and are therefore used on a wide scale as industrial materials (sheets, films, fibers, and the like) and as automotive and electrical/electronic parts, and are molded and processed by the numerous methods commonly employed for molding and processing plastics. However, very few practical results have been achieved concerning the application of calender molding (which is extensively used with vinyl chloride, ethylene/vinyl acetate copolymers (EVA), and rubber) to the molding and processing of sheets and films.

[0003] Calender molding is a process in which a resin is held in the gap between high-temperature calender rolls to perform plasticization, defoaming, and blending, and the resin is then calendered to the desired thickness in the roll gap. Polyester elastomers are often used as hot-melt adhesives, and are thus inherently disadvantageous in that their high tackiness causes them to excessively adhere or stick to the rolls when calendering is attempted. Raising the roll temperature in order to facilitate plasticization and defoaming tends to result in even higher tackiness, whereas lowering the roll temperature results in a slightly lower tackiness but makes it extremely difficult to separate and remove a film in a stable state from the rolls because polyester elastomers are crystalline resins with a clearly defined melting point, so plasticization deteriorates rapidly if the roll temperature is reduced below the optimum temperature in the vicinity of the melting point.

[0004] Attempts have been made to add polyolefin wax, pulverized fluororesins, aromatic amine compounds, pulverized silicone resins, or the like to polyester elastomers in order to address the aforementioned problems. Such methods can improve the tackiness and adhesion of the elastomers to the rolls, and studies have been conducted with the aim of putting these methods to practical use.

[0005] According to another known method, a polyvinyl chloride graft polymer is blended with a polyester elastomer, and the product is used in calendering applications (JP (Kokoku) 58-56545).

[0006]

**[Problems Which the Invention Is Intended to Solve]** To improve calendering, it is necessary, however, not only to control the tackiness and adhesion of the molten composition but also to prevent a so-called plate-out phenomenon, in which a layered deposit forms on the roll surface. Although the actual nature of the deposit formed by the plate-out phenomenon has not yet been definitively established, it is believed to result from the hardening of gelled products and other materials formed by the thermal degradation of the molten composition.

[0007] The conventional methods described above can result in improved initial tackiness and adhesion but sometimes fail to prevent the plate-out phenomenon from occurring during repeated operations, and hence to prevent stable commercial operations from being disrupted. An object of the present invention is to provide a polyester elastomer composition designed for calendering and endowed with improved plate-out resistance.

[0008]

**[Means Used to Solve the Above-Mentioned Problems]** Specifically, the present invention provides a polyester elastomer composition for calendering obtained by melting and admixing a phosphorus-based compound in an amount of 0.001–10 weight parts per 100 weight parts of a polyester block copolymer containing as principal constituent components thereof (A) high-melting crystalline polymer segments and (B) low-melting polymer segments comprising aliphatic polyether units and/or aliphatic polyester units.

[0009] The high-melting crystalline polymer segments (A) of the polyester block copolymer used in the present invention are composed of polyesters formed by (a) dicarboxylic acids and ester-forming derivatives thereof, and (b) diols and ester-forming derivatives thereof. The following compounds are preferred as dicarboxylic acids (a): terephthalic acid, isophthalic acid, phthalic acid, naphthalene-2,6-dicarboxylic acid, naphthalene-2,7-dicarboxylic acid, anthracene dicarboxylic acid, diphenyl-4,4'-dicarboxylic acid, diphenoxylethane dicarboxylic acid, 4,4'-diphenyl ether dicarboxylic acid, 5-sulfoisophthalic acid, sodium 3-sulfoisophthalate, and other aromatic dicarboxylic acids; 1,4-cyclohexane dicarboxylic acid, cyclopentane dicarboxylic acid, 4,4'-dicyclohexyl dicarboxylic acid, and other aliphatic dicarboxylic acids; and adipic acid, succinic acid, oxalic acid, sebacic acid, dodecanedioic acid, dimer acid, and other aliphatic

dicarboxylic acids. Ester-forming derivatives of these dicarboxylic acids, such as lower alkyl esters, aryl esters, carbonate esters, and acid halides, can also be used in the same manner.

Preferred examples of diols (b) include diols with a molecular weight of 400 or less, such as 1,4-butanediol, ethylene glycol, trimethylene glycol, pentamethylene glycol, hexamethylene glycol, neopentyl glycol, decamethylene glycol, and other aliphatic diols; 1,1-cyclohexane dimethanol, 1,4-dicyclohexane dimethanol, tricyclodecane dimethanol, and other alicyclic diols; and xylylene glycol, bis(*p*-hydroxy)diphenyl, bis(*p*-hydroxyphenyl)propane, 2,2-bis[4-(2-hydroxyethoxy)phenyl]propane, bis[4-(2-hydroxy)phenyl]sulfone, 1,1-bis[4-(2-hydroxyethoxy)phenyl]cyclohexane, 4,4'-dihydroxy-*p*-terphenyl, 4,4'-dihydroxy-*p*-quarter-phenyl, and other aromatic diols. These diols may also be used in the form of ester-forming derivatives such as acetyls and alkali metal salts.

[0010] These dicarboxylic acids, derivatives thereof, and diol components may also be used as combinations of two or more compounds. A polybutylene terephthalate derived from terephthalic acid or dimethyl terephthalate and 1,4-butanediol is the most preferred example of segments (A).

[0011] The low-melting polymer segments (B) of the polyester block copolymer used in the present invention are composed of an aliphatic polyether and/or aliphatic polyester. Examples of suitable aliphatic polyethers include poly(ethylene oxide)glycol, poly(propylene oxide)glycol, poly(tetramethylene oxide)glycol, poly(hexamethylene oxide)glycol, copolymers of ethylene oxide and propylene oxide, ethylene oxide addition polymers of poly(propylene oxide)glycol, and copolymers of ethylene oxide and tetrahydrofuran. Examples of suitable aliphatic polyesters include poly( $\epsilon$ -caprolactone), poly(enantolactone), poly(caprylolactone), and polybutylene adipate. Of these aliphatic polyethers and/or aliphatic polyesters, the following are preferred because of considerations related to the elastic characteristics of the resulting polyester block copolymer: poly(tetramethylene oxide)glycol, ethylene oxide adducts of poly(propylene oxide)glycol, poly( $\epsilon$ -caprolactone), polybutylene adipate, and the like.

[0012] The low-melting polymer segments (B) of the polyester block copolymer used in the present invention should be copolymerized in an amount of 10–90 wt%, and preferably 15–75 wt%. In particular, it is undesirable for this amount to be less than 10 wt% or greater than 90 wt%, because in the first case the product has insufficient flexibility or rubber elasticity, and in the second case low crystallinity results, and the moldability tends to decline.

[0013] The phosphorus-based compound used in the present invention, while not subject to any particular limitations, should preferably be one or more compounds selected from the compounds expressed by General Formulas (I), (II), and (III) below.

[0014]

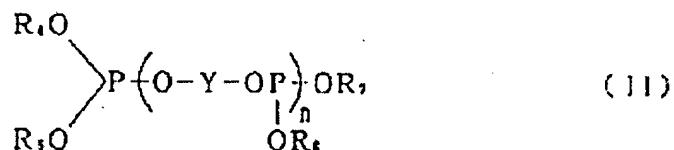
[Chemical Formula 4]



(In Formula (I), R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each a hydrogen atom; a group containing a C<sub>1-60</sub> substituted or unsubstituted alkyl, alkenyl, aryl, polyoxyalkylene alkyl ether, or polyoxyalkylene aryl ether; or one of these substituted groups; wherein the R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> may be bonded to each other with or without the aid of substituents.)

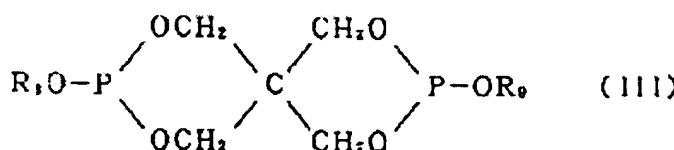
[0015]

[Chemical Formula 5]



[0016]

[Chemical Formula 6]



(In Formulas (II) and (III), R<sub>4</sub>-R<sub>9</sub>, which may be the same or different, are each a group containing hydrogen, alkyl, aryl, cycloalkyl aryl, alkyl aryl, aryl alkyl, alkenyl, aryl alkenyl, or

ether bonds; one of these groups containing an epoxy; or one of these substituted groups. Y is a polyhydric alcohol or polyhydric phenol residue. n is 0 to 10.)

Examples of the phosphoric acid compounds expressed by Formula (I) include dioctyl monophenyl phosphate, dicresyl monophenyl phosphate, xylenyl diphenyl phosphate, di-*t*-butyl phenoxy polyethoxy phosphate, lauryloxy polyethoxy phosphate, nonyl phenyl polyoxyethylene phosphate, tridecyl polyoxyethylene phosphate, oleyloxy polyethoxy dilauryl phosphate, and di(lauryloxy polyethoxy)phosphate.

[0017] Examples of phosphorous acid compounds expressed by Formulas (II) and (III) above include triphenyl phosphite, tridecyl phosphite, monophenyl didecyl phosphite, tricresyl phosphite, trisnonyl phenyl phosphite, tris(cyclohexyl)phenyl phosphite, tris(dipropylene glycol)phosphite, diphenyl phosphite, mono-2-ethyl hexyl phosphite, tetra(tridecyl)4,4-isopropylidene diphenyl phosphite, tetra(phenyl)dipropylene glycol diphosphite, bis(di-*t*-butyl phenyl)diphosphite, distearyl pentaerythritol diphosphite, heptakis(nonyl phenyl)tetrakis(bisphenyl A)pentaphosphite, heptakis(dipropylene glycol)triphosphite, bis(neopentyl glycol)dipropylene glycol diphosphite, tetrakis(butoxyethoxyethyl)ethylene glycol diphosphite, bis(cyclohexyl phenyl), and bis(phenyl ethyl)1,4-dibutane diol diphosphite. It is also possible to use metal salts in which these compounds are used as counterions. One, two, or more of these phosphorus compounds may be used in a ratio of 0.001–10 weight parts, preferably 0.01–5 weight parts, and ideally 0.05–3 weight parts, per 100 weight parts of the polyether ester elastomer.

[0018] According to the present invention, polyolefin wax should preferably be admixed in an amount of 0.001–10 weight parts per 100 weight parts of the polyester block copolymer. The polyolefin wax should preferably be a polyolefin with a number-average molecular weight of 50,000 or less, and it is not subject to any particular limitations as long as it imparts a waxy feel. Manufacturing methods include those in which an  $\alpha$ -olefin is polymerized as a starting material at elevated or reduced pressure, and those in which a product with a reduced molecular weight is obtained by decomposing a polyolefin previously polymerized to a high molecular weight. Another preferred product is a so-called modified polyolefin wax obtained by adding unsaturated groups, introducing carboxylic acid groups or hydroxyl groups into a molecule by oxidation or the like, or initiating a reaction that involves maleic acid or a compound containing epoxy groups. Examples of  $\alpha$ -olefins as starting materials for polyolefin wax include ethylene, propylene,

1-butene, 1-hexene, and 4-methylpentene. Two or more  $\alpha$ -olefins may be used as such starting materials, and two or more types may be used together. Preferred among these are polyethylene wax, polypropylene wax, and polybutylene wax with a molecular weight of 30,000 or less, and particularly preferred is a modified polyethylene wax with a molecular weight of 10,000 or less and an acid value (or hydroxyl value) of 10 or greater, as measured by the method defined in JIS K5902. It is unsuitable for the polyolefin wax to have a molecular weight of 50,000 or greater because this type of molecular weight reduces compatibility with the polyester elastomer and tends to have an adverse effect on the surface appearance of the molded article.

[0019] No particular restrictions are imposed on the method for adding the aforementioned compounds to the polyester block copolymer, and the following methods may be employed: methods in which the compounds are admixed into the polyester block copolymer in the molten state immediately after the reaction; methods in which the compounds are added to the resulting polyester block copolymer chips, and the addition product is heated, melted, and mixed; methods in which the compounds are dry-blended with polyester block copolymer chips immediately before calender molding, and the product is introduced into the calender molding process; methods in which the compounds are added to a polyester block copolymer in the form of a molten or semi-molten sheet during calender molding; and the like. When the compounds are melted and added to the resulting polyester block copolymer, the temperature thereof should be kept between the level 10°C below the melting point of the polyester block copolymer and 280°C. In addition, the mixing time in this case varies with the mixing temperature or mixing method between several seconds and 120 minutes.

[0020] The following compounds may also be arbitrarily added to the composition of the present invention as long as the objects of the present invention are not compromised: known antioxidants based on hindered phenols, thioethers, and the like; weather resistance agents based on benzophenols, hindered amines, and the like; epoxy compounds, isocyanate compounds, and other thickeners; silicone oil, metal stearates, metal montanates, montanic acid ester wax, and other mold release agents; dyes, pigments, and other colorants; titanium oxide, carbon black, and other UV absorbers; glass fibers, carbon fibers, potassium titanate fibers, and other reinforcing agents; silica, clay, calcium carbonate, calcium sulfate, glass beads, and other fillers; and talc and other nucleators; as well as fire retardants, plasticizers, expanding agents, fluorescents, preservatives/bactericides, cross-linking agents, and surfactants.

[0021] These additives or fillers may be admixed into the polyester block copolymer in advance, admixed into the polyester block copolymer together with the composition of the present invention, or added after the composition of the present invention has been molded.

[0022]

**[Working Examples]** The merits of the present invention will now be described through working examples. Unless indicated to the contrary, the terms "%" and "parts" used in the working examples are weight-based in all cases. The physical properties referred to in the working and comparative examples were measured in the following manner.

Melting point: DSC method

Hardness (Shore D scale): Measured according to JIS K-7215

Melt viscosity index (MFR value): Measured at a temperature of 200°C and a load of 2160 g in accordance with ASTM D-1238

Calender molding test: Starting material was introduced between rolls, and the time needed for the material to adhere to the rolls (roll smoothness preservation time) was measured. Two 8-inch rolls manufactured by Nippon Roll were used. The material was prepared by the dry blending of polyester block copolymer pellets and a specific calendering improver. In some cases, the material was prepared by melting and mixing the ingredients in advance and pelletizing the resulting mixture.

Plate-out: Roll contamination was visually monitored and graded on a five-point scale when the aforementioned calender molding tests were continuously performed for 20, 60, and 180 minutes on each sample without washing the rolls. "5" was assigned to cases that were considered satisfactory and did not have any plate-out at all, "4" was assigned to cases that were slightly inferior to "5" but still presented no problems, "3" was assigned to cases in which the roll surface luster decreased and long-term operation was believed to be impaired, "2" was assigned to cases with pronounced roll surface clouding, and "1" was assigned to cases in which severe plate-out occurred.

Blooming: A sheet with a thickness of 500  $\mu\text{m}$  that underwent the aforementioned calender molding test was allowed to stand at room temperature for 90 days or longer, and the surface condition of the sheet was visually monitored.

Tensile break elongation: Measured at a distortion factor of 400%/min with the aid of a 45% piece (JIS No. 2) clipped from a sheet with a thickness of 500  $\mu\text{m}$  obtained in the aforementioned calender molding test.

#### [0023] Reference Example

##### Polymerization of Polyester Elastomer (A-1)

Dimethyl terephthalate (194 parts), poly(tetramethylene oxide)glycol (265.4 parts) with a number-average molecular weight of about 1000, and tetramethylene glycol (69.6 parts) were introduced together with 0.10 part of titanium tetrabutoxide into a reaction vessel fitted with helical-ribbon stirring vanes. The system was kept for 2 hours at 190–225°C, and methanol was distilled out of the system in an amount equal to 95% of the stoichiometric methanol amount. Irganox 1010 (heat-resistant agent based on a hindered phenol and manufactured by Ciba-Geigy) was added in an amount of 0.5 part to the reaction mixture, the system was heated to 245°C, the pressure in the system was subsequently reduced to 0.2 mm Hg over a period of 50 minutes, and polymerization was performed for 2 hours under these conditions. The resulting polymer was ejected in strands into water, and the strands were cut, yielding pellets. This polymer was designated "A-1."

#### [0024] Polymerization of Polyester Elastomer (A-2)

Terephthalic acid (100 parts), 1,4-butanediol (110 parts), and tetrabutyl titanate (0.1 part) were introduced into a reaction vessel fitted with a rectifying column and helical-ribbon stirring vanes; reaction water was distilled out under stirring; an esterification reaction was conducted for 2 hours in a nitrogen atmosphere under normal pressure; the reaction product was transferred to a polymerization kettle; the pressure inside the system (250°C) was reduced to 0.2 mm Hg; and polymerization was performed for 2 hours under these conditions. The resulting polymer was ejected in strands into water, and the strands were cut, yielding polybutylene terephthalate with a relative viscosity of 1.47 and a melting point of 225°C. The polybutylene terephthalate and  $\epsilon$ -caprolactone were fed at the respective rates of 900 g/hr and 1700 g/hr to the hindermost feed port of a single-screw extruder (inside diameter: 30 mm, L/D: 40) in which a blending unit with a length of 200 mm was provided to the middle and distal sections, and an addition polymerization reaction was conducted while the rotational speed of the screw was kept at 30 rpm and the temperature of the middle section of the cylinder was set to 240°C. The polymer was subsequently ejected in strands from a die, and the strands were cut, yielding pellets. The pellets

(100 parts) and triphenyl phosphine (0.1 part) were blended using a vented single-screw extruder equipped with a full-flight screw (inside diameter: 30 mm, L/D: 40) while the degree of vacuum of the vent hole was kept at 10 mm Hg, the extrusion temperature was maintained at 200°C, and the rotational speed of the screw was set to 60 rpm. The  $\epsilon$ -caprolactone was removed, the catalyst was deactivated, the polymer was ejected in strands from a die, and the strands were cut, yielding pellets. These pellets were designated "A-2." Table 1 shows the compositions and physical properties of A-1 and A-2.

[0025]

**[Table 1]**

	Amount of copolymerized poly(tetramethylene oxide)glycol (wt %)	Amount of copolymerized polycaprolactone (wt %)	Melting point (°C)	Shore D hardness	Melt viscosity index (g/10 min)
A-1	65		165	38	8
A-2		65	171	39	26

### **Working Examples 1-6**

A product obtained by adding 1.2 weight parts of anatase-type titanium dioxide (white pigment) to 100 weight parts of the polyester elastomer A-1 obtained in the reference example was fashioned into a base mix, the (B-1) through (B-6) shown in Table 2 were dry-blended as examples of phosphorus compounds in the ratios shown in Table 3, and the aforementioned calender molding tests were performed at a roll temperature of 165°C. Table 2 shows the compounds used, and Table 3 shows the evaluation results of Working Examples 1-6.

### **Comparative Example [1]**

A product obtained by adding 1.2 weight parts of anatase-type titanium dioxide (white pigment) to 100 weight parts of the polyester elastomer A-1 obtained in the reference example was evaluated in the same manner as in Working Example 1. Table 3 shows the evaluation results of Comparative Example 1.

### **[0026] Working Examples 7 and 8**

A product obtained by adding 1.2 weight parts of titanium oxide (white pigment) to 100 weight parts of the polyester elastomer A-1 obtained in the reference example was fashioned into a base mix, the (B-1) shown in Table 2 was dry-blended as an example of a phosphorus compound in the ratio shown in Table 3, and the aforementioned calender molding tests were

performed at a roll temperature of 165°C. Similarly, the (B-3) shown in Table 2 was dry-blended as an example of a phosphorus compound in the ratio shown in Table 3, and the aforementioned calender molding tests were performed at a roll temperature of 165°C. Table 2 shows the compounds used, and Table 3 shows the evaluation results of Working Examples 7 and 8.

### **Comparative Examples 2 and 3**

Polyethylene wax (C-1) and (C-2) was dry-blended in the ratios shown in Table 3 with a product obtained by adding 1.2 weight parts of titanium oxide (white pigment) to 100 weight parts of the polyester elastomer A-1 obtained in the reference example, and the aforementioned calender molding tests were performed at a roll temperature of 165°C. Table 3 shows the evaluation results.

[0027]

[Table 2]

Additive	Structural formula	Remarks
B-1		—
B-2		—
B-3		—
B-4		—
B-5		—
B-6		—
C-1		Polyethylene wax Comex <sup>1</sup> 2000 by Sanyo Chemical Industries
C-2		Polyethylene wax Hi-Wax 110P by Mitsui Petrochemical Industries

<sup>1</sup> Translator's note: Tentative transliteration of a trade name that could not be found in the reference materials available to the translator.

[0028]

[Table 3]

Example	Polymer	Additive: phosphorus compound	Additive: other	Additive (weight parts)	Roll smoothness preservation time	Plate-out, 20 min	Plate-out, 60 min	Plate-out, 180 min	Blooming	Tensile break elongation (kg/cm <sup>2</sup> )	Elongation (%)
WE 1	A-1	B-1		0.6	20	5	5	5	5	280	1120
WE 2	A-1	B-2		0.6	23	5	5	4	5	284	1090
WE 3	A-1	B-3		0.6	38	5	5	5	5	307	1150
WE 4	A-1	B-4		0.6	36	5	5	5	5	304	1170
WE 5	A-1	B-5		0.6	31	5	4	4	4	299	1160
WE 6	A-1	B-6		0.6	25	5	5	5	4	275	1130
WE 7	A-1	B-1	C-1	0.6	60 or greater	5	5	5	5	285	1200
WE 8	A-1	B-3	C-2	0.6	60 or greater	5	5	5	4	287	1190
CE 1	A-1	—	—	—	1 or less					Sheeting impossible	
CE 2	A-1		C-1	0.3	35	3	1	1	1	4	287
CE 3	A-1		C-2	0.3	20	2	1	1	3	270	1090

WE: Working Example, CE: Comparative Example

### [0029] Working Examples 9–11

The same experiments and evaluations as in Working Examples 1–8 were performed except that A-2 and A-3 were used instead of the polyester elastomer A-1 obtained in the reference example. For A-2, the calender molding tests were performed at a roll temperature of 170°C. The results are shown in Table 4.

### Comparative Examples 4 and 5

Polymer A-2 of the reference example was evaluated in the same manner as in Working Example 9. The results are shown in Table 4.

[0030]

[Table 4]

Example	Polymer	Additive: phosphorus compound	Additive: other	Additive (weight parts)	Roll smoothness preservation time	Plate-out, 20 min	Plate-out, 60 min	Plate-out, 180 min	Blooming	Tensile break elongation Strength (kg/cm <sup>2</sup> )	Elongation (%)
WE 9	A-2	B-1		0.6	22	5	4	4	4	290	1080
WE 10	A-2	B-3		0.6	30	5	4	4	4	285	1060
WE 11	A-2	B-1	C-1	0.6 0.3	60 or greater	5	4	4	5	286	1100
CE 4	A-2	—	—	—	1 or less					Sheeting impossible	
CE 5	A-2		C-1	0.3	31	2	1	1	4	277	1070

WE: Working Example, CE: Comparative Example

### [0031] Working Examples 12 and 13

The same evaluation was performed using products obtained by a process in which either (B-1) or (B-1) and (C-1) were selected from the additives of Working Examples 1–6 and mixed in advance in the ratios shown in Table 5 in a V-blender with the polyester elastomer A-1 obtained in the reference example, and the product was melted, blended, and pelletized at 190°C with the aid of a twin-screw extruder fitted with a screw having a diameter of 45 mm. The evaluation results of Working Examples 12 and 13 are shown in Table 5.

[0032]

[Table 5]

Example	Polymer	Additive: phosphorus compound	Additive: other	Additive (weight parts)	Roll smoothness preservation time	Plate-out, 20 min	Plate-out, 60 min	Plate-out, 180 min	Blooming	Tensile break elongation Elongation (%)
WE 12	A-1	B-1		3.0	45	5	5	5	4	280 1100
WE 13	A-1	B-2	C-1	2.5 0.5	60 or greater	4	5	5	4	264 1020

WE: Working Example

The results in Table 3 indicate that the composition of the present invention can be endowed with dramatically improved copolymers, and particularly plate-out resistance, by the addition of compounds in low proportions, that excellent melting points can be obtained, and that the same excellent results can be obtained by the combined use of phosphorus compounds and polyolefin waxes.

[0033] It is also apparent from the results shown in Table 6<sup>2</sup> that roll smoothness and plate-out resistance can be appropriately improved by admixing the aforementioned compounds to A-2 and A-3, which are compounds whose soft segments differ from those of polyester elastomer A-1, and that the roll adhesion of these polyester elastomers is markedly improved irrespective of the type of soft segments in the polyester elastomers. In addition, the composition of the present invention yields the same results when the compounds are added in greater amounts, as can be concluded from the results shown in Table 4.

[0034]

**[Merits of the Invention]** The polyester elastomer composition for calendering obtained in accordance with the present invention is capable of yielding molded sheets or films that have adequate calendering properties, particularly plate-out resistance, as well as excellent surface appearance, mechanical properties, heat resistance during processing, and color tone during processing.

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(Continued from front page)

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<sup>2</sup> Translator's note: There is no Table 6 in the original.